

Effect of Recycled Denture Base Polymeric Powder Incorporation on the Surface Properties of Heat-cured PMMA Denture Base Acrylic Resin: An *In Vitro* Study

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ABSTRACT

Aim: The present *in vitro* research aimed to assess the polishability (P), translucency (T), surface hardness (SH), and surface roughness (SR) of heat-cured PMMA-based denture base resin processed after modification with recycled denture base resin (r-PMMA) at 10, 20, 30, 40, and 50% (w/w) to commercially available resin (R₁₀, R₂₀, R₃₀, R₄₀, and R₅₀) (w/w).

Materials and methods: In total, 90 rectangular specimens ($n = 15$) were fabricated to test P and T and were analyzed through visual inspection accordingly to ISO standardization. 90 disk-shaped specimens ($n = 15$) were fabricated to test SR and SH. The mean arithmetic roughness (R_a) was recorded using a surface profilometer to assess the SR. Vicker's microhardness testing was done to obtain Vickers hardness number (VHN). Data were tabulated and compared using ANOVA ($\alpha = 0.05$). Further *post hoc* Bonferroni tests were performed on each pair of groups.

Results: All the specimens tested for P and T comply with the ISO 1567. The R_a of the control group- SR R₀ was found to be 0.10 μm . The modification resulted in an increase in SR and was found to be ranging from 0.11 μm (SR R₁₀) to 0.16 μm (SR R₅₀). The VHN recorded for the control group (SH R₀) was 20.35 kg/mm². The experimental groups demonstrated a decrease in VHN, from 16.67 kg/mm² (SH R₁₀) to 13.85 kg/mm² (SH R₅₀).

Conclusion: The polymeric modification does not affect the P and T of the cured specimens. The modification resulted in an increase of SR proportional to the w/w of r-PMMA added. The experimental groups demonstrated a decrease in VHN as a function of an increase in the addition of r-PMMA w/w.

Clinical significance: The ability to reuse denture base resin would significantly reduce the nonbiodegradable type of biomedical waste that is produced and sent out of the dental institutes and practice.

Keywords: Acrylic resins, Hardness test, Recycling, Reuse.

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INTRODUCTION

Dental healthcare professionals have dedicated themselves to enhancing and promoting better oral health and hygiene. An array of treatment modalities and dental materials are employed to do so. With the advancements in material science, there has been hand-in-hand progress in contemporary dental materials. With greater awareness about the importance of oral health, the past century has seen a steady increase in the quality and quantity of dental material entering the market.

The long and illustrious history of materials used as denture bases started in the 18th century when ivory was used as a dental substitute. Japanese woodworkers carved dentures from sweet-smelling species of wood around the 8th century. With the dawn of the metal ages, various metals such as Gold, Silver, Stainless steel, and base metal alloys—Ni-Cr and Co-Cr were also utilized as denture base materials. Alexis Duchateau, a Parisian apothecary, even attempted to make dentures from porcelain along with Parisian dentist Nicholas Dubois de Chemant. But it was in the mid-1930s that a breakthrough invention was introduced in the form of acrylic type plastic denture base, Vernomite. By 1946, approximately 95% of all dentures were fabricated using PMMA, Wright conducted a clinical evaluation on methyl methacrylate and found that they fulfilled most requirements of ideal denture base material. Auto polymerizing resins were introduced after the development of chemical accelerators in Germany around 1947.¹⁻³

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The unparalleled popularity of PMMA and its copolymers can be attributed to its ideal properties such as biocompatibility, stability in the oral environment, ease of manipulation, ready availability, and optical properties. Different denture base materials like Vinyl acrylic

(Luxene 44), Polystyrene (Charles Dimmer), nylon, polycarbonate, polyethers, polysulphones, epoxy resin, high impact methacrylate, and Polypropylene were used from 1951–1967, but could not be used as absolute substitutes of PMMA. Ongoing research aims at improving the mechanical strength of PMMA-based denture base materials by incorporating various reinforcements/modifications to the polymer and/ or monomer components.

The increasing consumption of diminishing natural resources, air and water pollution, ever-growing landfills, and the effects of global warming, makes saving our environment imperative. The Ministry of Environment, Forest and Climate Change, Government of India reported that in 2018–2019 the Solid Waste Generation and Collection in India was 54,417.385 TPD and 45,082.15 TPD, respectively. The report also established that 15,386.81 TPD of solid waste was treated and 22,904.70 TPD ended in landfills. The annual plastic consumption was chalked up to nearly 8 million tons and the plastic waste generation per day was reported at approximately 25,940 tons in India. The annual report on biomedical waste reported that a total of 2,38,170 Health Care Facilities (HCFs) generated biomedical waste of 557 Tonnes per day in India.⁴

WHO reports on healthcare waste management status in countries of the South-East Asia Region in 2017, noted that 1000 tons/day and 35,000 tons/year of healthcare waste was produced by the 11 South Asian countries. It has been identified the inadequate and improper handling of HCW has contributed significantly to environmental pollution. Increasing global concerns have led WHO to recommend that countries should develop customized national plans for safe management of HCW based on data on the quantity and quality of biomedical waste generation through surveys. One of the first South Asian countries to establish legislation regarding biomedical waste management was India. Though dental health care providers also generate a substantial amount of biomedical waste, no specific provision was mentioned for the same. Dental health care professionals are yet to be provided with any specific guidelines, manuals, or protocols in relation to the management of dental healthcare waste.⁵

Today's commercially available heat cure denture base resin materials are supplied in 2-component powder-liquid system. The powder consists mainly of prepolymerized Poly (methyl methacrylate) and liquid component consisting of methyl methacrylate in monomeric form as the major component. Many researchers have discussed improvising the physical properties of the denture base resins by means of substitutions and modifications to both the monomer and polymer. With the world moving toward environmental consciousness, we as dental professionals must take up the responsibility to make our profession keep pace with time and move forward toward "Green dentistry."^{6,7}

The present research aims at evaluating the effect of recycled denture base polymeric powder incorporation on the surface properties of heat-cured PMMA denture base acrylic resin base resin (r-PMMA) at the concentration of 10, 20, 30, 40, and 50% (w/w) (R_{10} , R_{20} , R_{30} , R_{40} , and R_{50}) to commercially available heat cure denture base resin. The null hypothesis for the research was that the substitution will not affect the polishability (P), translucency (T), surface roughness (SR), and surface hardness (SH) of the modified cured denture base acrylic resin.

MATERIALS AND METHODS

Heat-polymerizing denture base acrylic resin (DPI Heat Cure, DPI, Mumbai, India) was modified by substitution with 10, 20, 30, 40,

and 50% (w/w) of recycled denture base resin (R_{10} , R_{20} , R_{30} , R_{40} , and R_{50}) of recycled denture base resin (r-PMMA) and used to fabricate experimental group specimens. The control group specimens were fabricated using unmodified denture base resin following the manufacturer's instructions (R_0). Flowchart 1 and Table 1 summarize the methodology followed and sample preparation for the research, respectively.

Specimen Preparation

Polishability and Translucency

A total of 90, $64 \times 40 \times 5$ mm rectangular specimens ($n = 15$ for each group) were fabricated accordingly to ISO 1567, by using metal dies of appropriate dimension to analyze P and T. Mold space was created using elastomeric addition silicone putty consistency material (Photosil Soft Putty (Addition Cure), DPI, Mumbai, India) and the prepared dies in the brass dental flask. The modified polymer was mixed with commercially available monomer and packed as the mixer reached the dough stage, in the mold space previously produced. The packing pressure of 3500 psi was applied for 10 minutes using a hydraulic press (P 400–Hydraulic press, Sirio Dental Division, Italy). This was followed by a long curing cycle of 8 hours at 74°C, followed by terminal boiling for 1 hour at 100°C in an acrylizer (Unident Instruments, New Delhi, India). Specimens were retrieved by deflasking after ½ hour of bench cooling. The retrieved specimens were ground using 30µm emery, followed by polishing with pumice of 10–20 µm for ≤1 minute and wet wheel and dry wheel. All polymerized specimens were maintained at 37°C for 48 ± 2 hours in distilled water before the commencement of testing, to allow for the leaching out of monomeric residues. The specimens were visually inspected according to ISO 1567:1999 for Dentistry–Denture Base Polymers.⁸

Surface Roughness and Surface Hardness

A total of 90 disk-shaped (20×3 mm²) specimens ($n = 15$) were fabricated to test SR and SH following a similar protocol as for P and T.^{9,10}

Testing

Polishability

Polishability was examined by visual inspection after the specimens were finished and polished according to the above-mentioned protocol. The specimens were examined according to ISO 1567:1999. The specimen was said to have passed the visual inspection if the specimen retained its form with defined edges after deflasking, that is, no visible distortion is perceived after processing. And the specimens had a smooth, hard, and highly glossy surface after the finishing and polishing protocol.⁸

Translucency

Each specimen was examined separately by positioning the finished and polished approximately 5 mm from a Frosted 40 W light bulb with an opaque disc (10×2 mm) centered on the surface nearest to the light bulb in a dark room. The specimen was said to be complying to the ISO 1567 standards when, from the opposite side of the specimen being tested, a shadow of the illuminated disc was.⁸

Surface Roughness

SR was analyzed using a profilometer (Surftest 211, Mitutoyo Corp., Tokyo) with a diamond stylus of 2 µm tip diameter (Fig. 1). SR is a two-dimensional parameter that was measured within a preset

Flowchart 1: Flowchart of the methodology

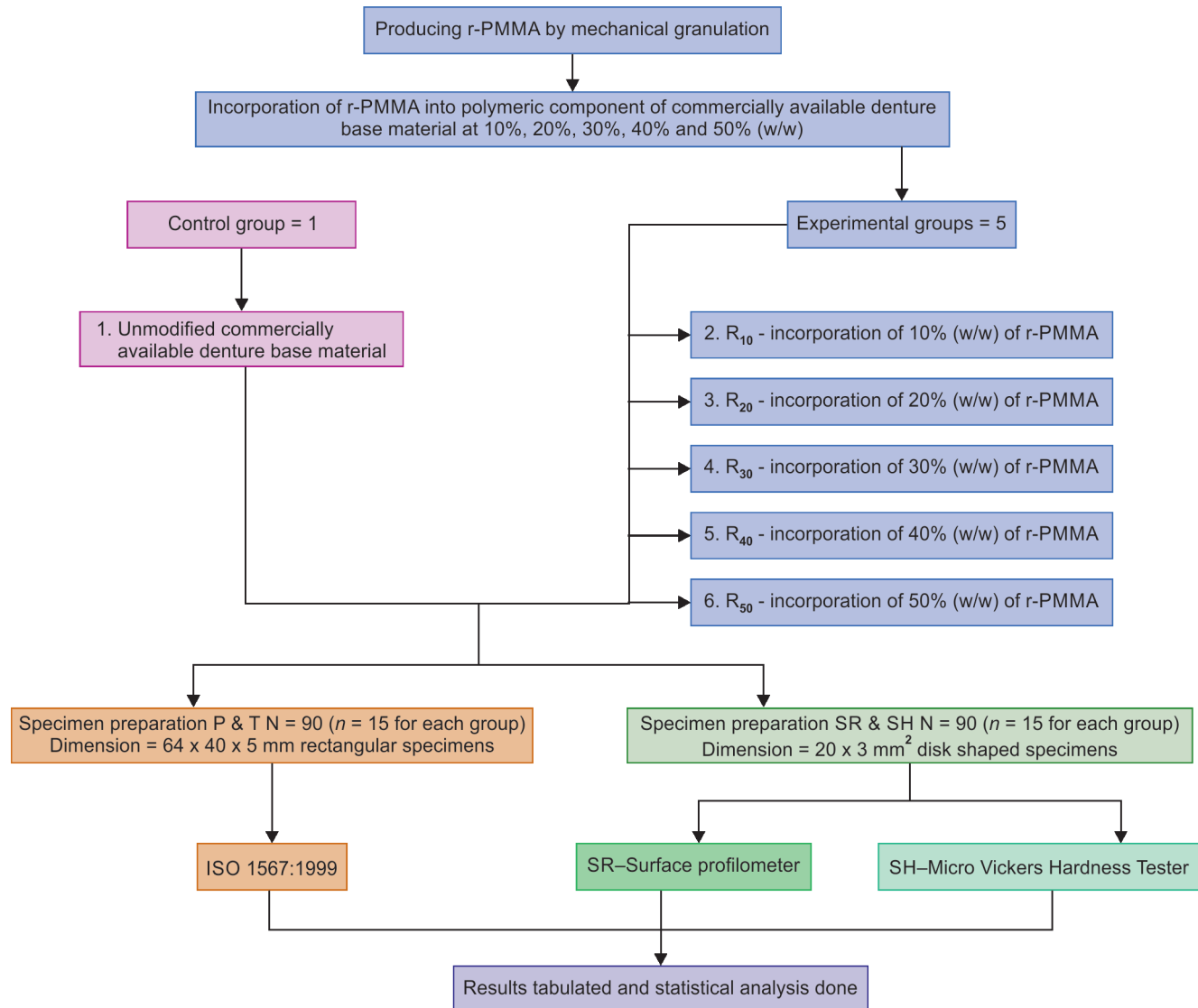


Table 1: Sample distribution

Group name	% of recycled denture base resin (w/w)	Sample size (n)	Sample code and sample dimension			
			P	T	SR	SH
			64 × 40 × 5 mm	64 × 40 × 5 mm	20 × 3 mm ²	20 × 3 mm ²
Control group R ₀	0	15	PR ₀	TR ₀	SR R ₀	SH R ₀
Experimental group R ₁₀	10	15	PR ₁₀	TR ₁₀	SR R ₁₀	SH R ₁₀
Experimental group R ₂₀	20	15	PR ₂₀	TR ₂₀	SR R ₂₀	SH R ₂₀
Experimental group R ₃₀	30	15	PR ₃₀	TR ₃₀	SR R ₃₀	SH R ₃₀
Experimental group R ₄₀	40	15	PR ₄₀	TR ₄₀	SR R ₄₀	SH R ₄₀
Experimental group R ₅₀	50	15	PR ₅₀	TR ₅₀	SR R ₅₀	SH R ₅₀

length on the specimen, as the heights of the surface irregularities. The mathematical average of this absolute value recorded is calculated and expressed as R_a . SR was measured by tracing three points randomly selected, for each specimen at 0.5 mm/s for 0.8 mm. The SR (R_a) was the specimen recorded as the mean of the three measurements obtained.

Vicker's Hardness Test

HMV-G31-FA, Micro Vickers Hardness Tester, (Shimadzu, Japan) was used to analyze the surface microhardness of the prepared samples (Fig. 2). The tester is equipped with a standardized automated length measurement function which used a built-in digital camera in its frame. The system was able to recognize the shape of the

specimens and automatically sets the test position. Each specimen was subjected to five indentations at random points with 50 gm of load for 15 seconds. The software automatically set magnification for the optimal lens from the anticipated hardness. The system overlapped the images obtained to create a complete image of the indentation and then measured the diagonal length of the indentation that the indenter made on the sample. The software automatically calculated and displayed the results in terms of the average of the five readings of the hardness, maximum value, minimum value diagonal line length, conversion value, standard deviation, coefficient of variation, of the Vicker's microhardness number (VHN) for each specimen.

Statistical Analysis

Statistical Package for the Social Sciences (SPSS) software program, SPSS Inc., Chicago, IL, USA, version 26.0 was used to Statistical analyze the results tabulated. To compare the mean between outcome variables between the groups, analysis of variance (ANOVA) was applied. When ANOVA test came significant, the *post hoc* Bonferroni test was applied for mean comparison between each group. The *p*-value was set at ≤ 0.05 for statistical significance.

RESULTS

All of the specimens prepared passed the P and T tests. The mean SR (R_a) and mean VHN of all the six groups tested are presented in Table 2 and Table 3, respectively. The mean R_a recorded for the

control group (SR R_0) was 0.10 μm , which was the least of all the six groups. The mean R_a for the five experimental groups, SR R_{10} , SR R_{20} , SR R_{30} , SR R_{40} , and SR R_{50} was recorded as 0.11 μm , 0.12 μm , 0.14 μm , 0.15 μm , and 0.16 μm , respectively. The highest mean R_a was recorded for the experimental group SR R_{50} . There was a significant difference in the mean SR between the groups indicated by a *p*-value of ≤ 0.05 (Table 2). The *post hoc* Bonferroni test indicated that the mean SR for groups SR R_0 and SR R_{10} , SR R_{10} and SR R_{20} , SR R_{30} , and SR R_{40} were similar (Table 4).

The mean VHN recorded for the control group (SH R_0) was 20.35 HV, which was the highest of all the six groups being tested. The mean VHN for the five experimental groups, SH R_{10} , SH R_{20} , SH R_{30} , SH R_{40} , and SH R_{50} was recorded as 16.67 VH, 16.46 VH, 16.02 VH, 15.81 VH, and 13.84 VH, respectively. The lowest mean VHN was recorded for the experimental group SH R_{50} . A significant difference in the recorded mean SH between the groups was noted (Table 3). The *post hoc* Bonferroni test indicated that the mean SH for groups SH R_0 and SH R_{10} , SH R_{10} , SH R_{30} , SH R_{20} , and SH R_{30} , SH R_{20} and SH R_{30} , SH R_{30} and SH R_{40} were similar (Table 5).

Table 2: Comparison of mean surface roughness of the different groups

Group	Mean	SD	F-value	p-value
SR R_0	0.10	0.01	69.05	0.001
SR R_{10}	0.11	0.01		
SR R_{20}	0.12	0.01		
SR R_{30}	0.14	0.01		
SR R_{40}	0.15	0.01		
SR R_{50}	0.16	0.01		

Table 3: Comparison of mean surface hardness of the different groups

Group	Mean	SD	F-value	p-value
SH R_0	20.35	1.09	122.47	0.001
SH R_{10}	16.67	0.47		
SH R_{20}	16.46	0.59		
SH R_{30}	16.02	0.57		
SH R_{40}	15.81	0.54		
SH R_{50}	13.84	0.95		

Table 4: *Post hoc* Bonferroni test to find out the difference in mean surface roughness between the groups

Group 1	Group 2	Mean difference	p-value
SR R_0	SR R_{10}	0.008	0.765
	SR R_{20}	0.015	0.007
	SR R_{30}	0.036	0.001
	SR R_{40}	0.049	0.001
	SR R_{50}	0.064	0.001
SR R_{10}	SR R_{20}	0.007	1
	SR R_{30}	0.027	0.001
	SR R_{40}	0.041	0.001
	SR R_{50}	0.055	0.001
	SR R_{30}	0.021	0.001
SR R_{20}	SR R_{40}	0.033	0.001
	SR R_{50}	0.048	0.001
	SR R_{30}	0.013	0.057
SR R_{30}	SR R_{50}	0.028	0.001
	SR R_{40}	0.015	0.011



Fig. 1: Profilometer being used to analyze the surface roughness (R_a) (screen showing the digital measurement of surface roughness of the specimen being displayed)

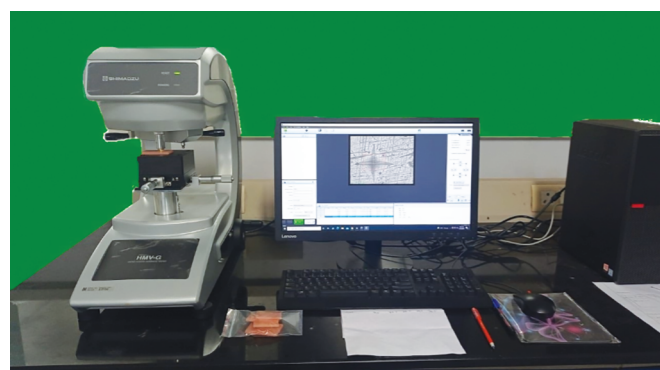


Fig. 2: Micro Vickers Hardness Tester (screen showing the digital image of the indentation produced being measured)

Table 5: *Post hoc* Bonferroni test to find out the difference in mean surface hardness between the groups

Group 1	Group 2	Mean difference	p-value
SR R ₀	SR R ₁₀	3.68	0.001
	SR R ₂₀	3.88	0.001
	SR R ₃₀	4.32	0.001
	SR R ₄₀	4.54	0.001
	SR R ₅₀	6.51	0.001
SR R ₁₀	SR R ₂₀	0.21	1
	SR R ₃₀	0.64	0.298
	SR R ₄₀	0.86	0.03
	SR R ₅₀	2.82	0.001
SR R ₂₀	SR R ₃₀	0.44	1
	SR R ₄₀	0.66	0.256
	SR R ₅₀	2.61	0.001
SR R ₃₀	SR R ₄₀	0.22	1
	SR R ₅₀	2.17	0.001
SR R ₄₀	SR R ₅₀	1.95	0.001

DISCUSSION

Literature is scarce on the quantification of solid waste and its management in dental scenarios. Research conducted by M Ozbek at a Dental College in Turkey suggests that the highest weight of waste came from the Prosthodontics Clinic where most dentures and impression materials are constructed and possibly disposed of (data shows over 30% of the total weight on the average) is expected to contribute to higher quantities due to the relatively high weight of these materials.¹¹ According to investigations by K Pushpanjali, it was estimated that 41,535 kg and 8307 kg of infectious and recyclable waste was generated by all the registered private clinical practices in Bengaluru city, India. The study also highlighted poor management practices of lead foil, gypsum products, and a lack of recycling practices.¹²

It was reported in 2013 by the dental services of Universitas Padjadjaran Dental Hospital (RSGM Unpad), one of the 27 dental colleges in Indonesia, that the use of dental alginate impression can reach 900 kg per month. According to a study by Wulandari on the dumping of medical waste in individual dentistry in Bandung, waste derived from dental alginate molds was disposed of in the garbage along with other infectious waste, and further disposal was united with domestic waste. This suggests that dental alginate mold waste was treated improperly and not recycled.¹³

Efforts to reuse dental alloys used for casting have been documented for decades. These studies concentrate on the predicting optimum number and percentage of reuse for producing restoration which can function intraorally without compromise.^{14–18} Numerous studies report efforts taken by orthodontics to condition and reuse orthodontic brackets and wires in order to reduce wastage.^{19–22} Researchers have also sought to recycle and reuse the Zirconia waste generated through CAD-CAM milling to produce CAD-CAM blocks for reuse in dentistry.²³ Gypsum products and investment materials have been suggested for various reuse without need for chemical or physical modification in areas like agriculture as compost additive, to reduce toxin runoff, in animal beddings and so on.^{24,25}

A study by T Frahdian et al. on Dental alginate impression material was aimed to remedy the above situation and to discover

its effects as additional fertilizer on the plant yields by determining the weight of cauliflower crop, and toward the quality of soil by determining the soil pH (Ultisol® Jatinangor). The team concluded that the addition of dental alginate waste at the dose of 0.01% and 0.1% increased the weight of cauliflower, but has no significant effect on the soil quality.¹³ Studies focusing on the recycling or reuse of acrylics used in dentistry are alarmingly scarce.²⁶ None have reported recycling denture base resins.

The dentures constructed at the dental institutes and clinics are composed of acrylic resin–Polymethyl methacrylate (PMMA). Most of the plastic-type waste along with used denture base material is placed in the red color-coded containers intended for recycling and sent to registered recyclers. PMMA is one of the most used thermoplastics due to its high quality and optical properties (transparent all-weather sheets, electrical insulation, automotive parts, surface coating, ion exchange resins, etc.).

It is worth mentioning that according to the Dental Council of India, India has a total 2,80,290 registered dentists, with 276 and 315 dental institutes offering master's and bachelor's courses in dentistry.²⁷ Heat cure denture resin (PMMA) usage has only increased in dental teaching institutes with an increase in graduate intake every year.

Various authors have attempted to improvise the mechanical-physical and chemical properties of the PMMA-based denture base resins. Both monomeric and polymeric modifications are common. To the best of the author's knowledge, there is no literature to be found attempting to recycle and reuse denture base resin, as polymeric incorporation. This first of its kind research aimed at evaluating the effect of polymeric modification done by incorporating recycled denture base resin (r-PMMA) at 10, 20, 30, 40, and 50% (w/w) (R₁₀, R₂₀, R₃₀, R₄₀, and R₅₀) (w/w) to the commercially available denture base resin, on the following surface properties: P, T, SR, and SH.

Literature has shown that surface properties like P and SR have a strong correlation with plaque accumulation and *Candidia albicans* colonization on the denture's surfaces. Clinically acceptable SR threshold levels reported by Bollen CM et al. is $\leq 0.2 \mu\text{m}$, below this critical level, there was no significant impact on intraoral plaque retention and plaque accumulation on restorative and prosthetic materials.²⁸

SR is a two-dimensional parameter that determines microbial adherence, biofilm formation, and colonization. An increase in SR can provide for areas for plaque and food debris collection.^{29–34} After studying the plaque retention and accumulation process from 3–6 days on prosthetic and restorative materials with various SR and surface energy, Nassar et al. concluded that plaque accumulation is more on a rough surface than on the smooth surfaces.³⁵ Also making the cleaning of denture surface difficult by regular hygiene methods. It was suggested by Quirynen M and Bollen CM that a smooth surface was paramount for denture hygiene and that roughness $\geq 2 \mu\text{m}$ would result in an exponential increase in bacterial retention on the surface.^{28,36} This finding was supported by Williams DW and Lewis MAO.³⁷ A review of the literature shows that a well-polished denture base surface has an SR of approximately $0.12 \mu\text{m}$, which is below the suggested critical level in literature.³⁸

In the current research, SR was calculated using a profilometer, yielding numerical data, the R_s value of the material as suggested by the literature.^{29–33} The SR of SR R₀ (control group) was found to be $0.10 \mu\text{m}$, which is analogous to the results by Abuzar et al. and Chatzivasilieiou et al., where conventional finishing and polishing techniques were used on heat-cured denture base resin.^{39,40} The SR values reported by Ulusoy et al. and Zissis et al. were found to

be much higher, this may be attributed to the fact that no finishing process was carried out on the specimens.

The modification resulted in an increase of SR proportional to the w/w of r-PMMA added and was found to be ranging from 0.11 μm (SR R_{10}) to 0.16 μm (SR R_{50}). The highest SR values were recorded for the SR R_{50} experimental group, which contented 50% of the r-PMMA, this is close to the 2 μm critical value. This increase could be due to the presence of loosely attached clusters of r-PMMA which could have been detached easily during the process of finishing and polishing, leaving detectable voids. This may have resulted due to the mechanical grinding process followed to obtain the r-PMMA, further investigation is needed to ascertain the reason for the increase in SR of the experimental groups. Literature is lacking in evaluating the effect of polymeric modification of PMMA-based denture base resin on SR.

The SH of the denture is an eminent property influencing its intraoral tribological performance. An indenter made of a harder material is used to indent the surface of the material to measure its SH. This value could indicate the possible degeneration of the polymeric matrix and denture longevity.^{9,10} The presence of residual monomer in the cured resin may affect its mechanical properties, especially SH. Thus, previous studies have evaluated residual monomer content to indirectly assess the SH.⁴¹

Surface hardness was tested using a microhardness tester and recorded as Vickers hardness number (VHN). This research recorded the VHN of the control group (SH R_0) as 20.35 kg/mm,² which is analogous to that reported in the literature.⁴²⁻⁴⁵ The experimental groups demonstrated a decrease in VHN as a function of an increase in the addition of r-PMMA w/w, from 16.67 VH (SH R_{10}) to 13.85 VH (SH R_{50}). Hardness is found to be closely related to the amount of residual monomer in the cured resin.⁴⁶⁻⁴⁸ An increased residual monomer content is known to facilitate softening of the cured resin by acting as a plasticizer. This reduces the polymer interchain forces, leading to easy deformation of the material under load.⁴⁹

As this research is one of the first to utilize recycled PMMA as polymeric modification, comparing and contrasting the results is difficult, due to a lack of literature. This research has attempted to evaluate the effects of the modification on surface properties *in vitro*, future research is needed on the subject in a clinically simulated environment to substantiate the present results. Future inquiries may be carried out to evaluate other dimensions of the polymeric modification, to better understand the scope of the modification, before real-world application.

CONCLUSION

Under the conditions of this present research, it may be concluded that the polymeric modification of PMMA-based denture base resin using recycled PMMA, does not affect the P and T of the cured specimens. The modification increased the mean SR of the modified cured specimens as a function of the r-PMMA (w/w) being added. Though the increased values were clinically acceptable. A decrease in the Vickers microhardness number as a function of the r-PMMA (w/w) being added was also observed.

SOCIAL IMPLICATIONS

The recycling and reuse of dental materials is an area of focus for the sustainable growth of dentistry in the future. The ability to reuse a dental material will greatly reduce the burden on the environment and the nonrenewable natural resources which are used to produce these materials.

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